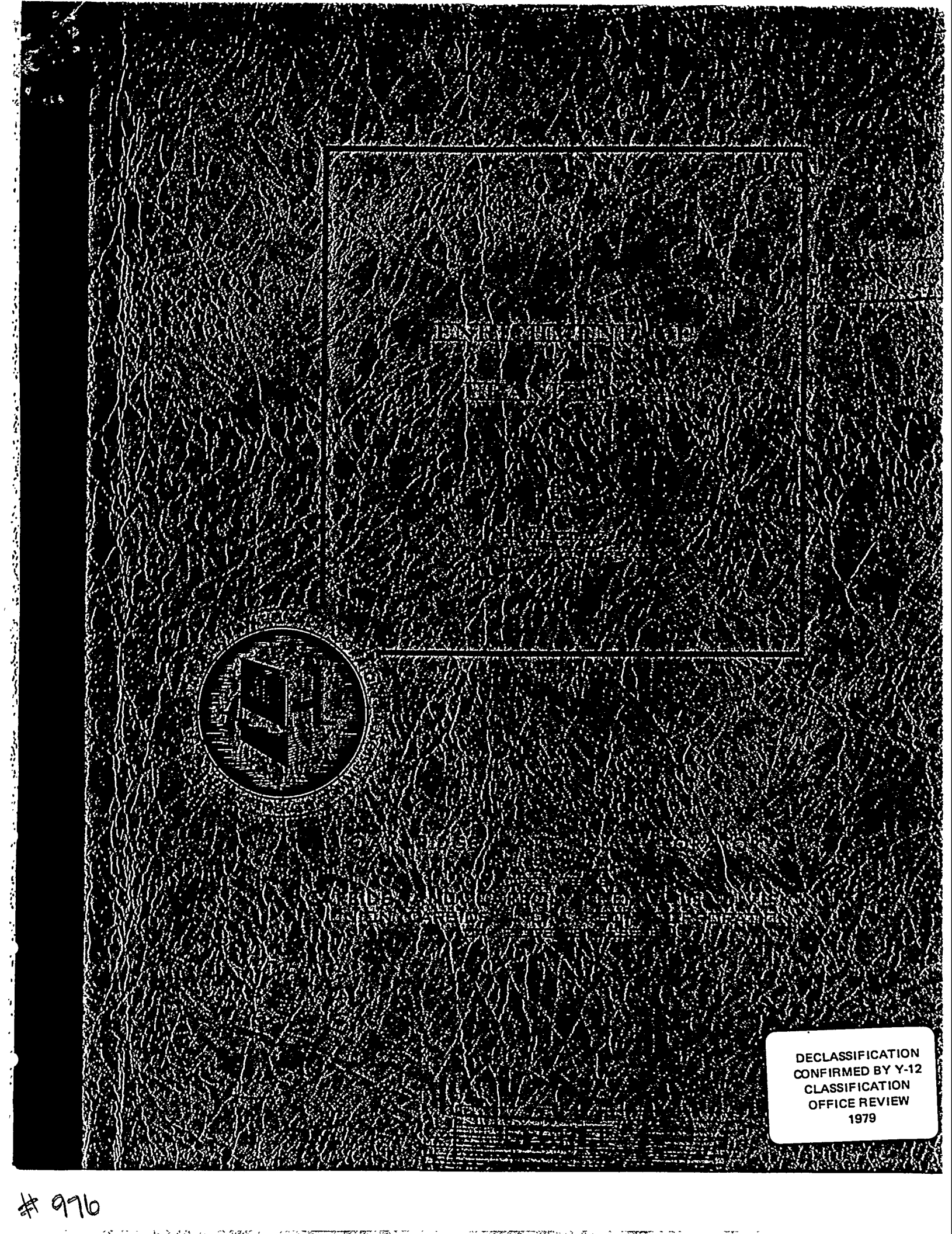

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Index No. Y-660
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This is copy 31 of 80, Series A.
Subject Category: Isotope Separation

ENRICHMENT OF URANIUM 234

B. Harmatz
R. S. Livingston

September 1, 1950

ELECTROMAGNETIC RESEARCH DIVISION
Dr. R. S. Livingston, Director

OAK RIDGE NATIONAL LABORATORY
Y-12 AREA
Operated by
CARBIDE AND CARBON CHEMICALS DIVISION
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Oak Ridge, Tennessee

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Incremental Costs.

Chemical Operators	③	@ 500	1500
Clerical Operators	④	@ 500	2000
Technician	④	@ 600	2400
Chemical Supv	1	@ 700	700
Track Supv	1	@ 700	700
~~Chemical~~			~~1000~~
1 Mechanic		500	500
1 E mechanic		500	500
1 General Mechanic		500	500
Misc. Labor			1000
			9000
			0.60

$\times 1,65 = 16500$

20,000 /mo.

24
1760
730
2760

$\frac{\quad}{8\ 760}$ = 27.40

6° Beam – 10 mg U/hr

n = 30, 200

10 mg/hr U

1.25% , 35% , = 36.25 total

63.75

$\frac{1.25}{35} \times 30 = 1.072 = \frac{n_{24}}{n_{25}}$ $\quad \frac{n_{24}}{n_{25}} = 1$ .933

$\frac{1.25}{63.75} \times 250 = 4.91$ $\quad \frac{n_{24}}{n_{28}} = 1$ .2037

2.137

$\frac{n_{24}}{n_{25}} + \frac{n_{24}}{n_{28}} + \frac{n_{24}}{n_{24}} =$

$\frac{n_{24}}{n_T} = \frac{1.072}{2.137} = 50.2$

10.22

46.80 43.70%

Same run 9. 26.1 205

30.

30 ma

– 32 ma

– 64 ma $\times 8.92 \times 1.25 =$ 7.14 mg/hr natural for two ares

(A) $\frac{50,000}{7.14} = 7,000$ hours. for a two are unit $\frac{\$25}{7.14} = \$3.5$/mg U^{234}

(B) $\frac{7.323}{762} = 9.60$ mg/hr U^{234}

$\frac{50,000}{9.60} = 5,210$ hours. for a two are unit $\frac{\$25}{9.60} = \$2.6$/mg U^{234}

$25

seven days a week

400 gram Charge.

CASE A. – AVERAGE OF U^{234} 1ST STAGE PERFORMANCE

1. Average U^{234} separation rate $\frac{7.323 \text{ gm}}{7.62 \times 10^3 \text{ h}} = 9.62 \times 10^{-3}$ g/h (two cores) for 1.34% feed level

2. Enrichment Factors. $U^{234} : U^{235} = 17.6$
$U^{234} : U^{238} = 162.0$

3. Process Efficiency : 9.7 percent

CASE B. – AVERAGE OF U^{234} 2nd STAGE PERFORMANCE

1. Average U^{234} separation rate $\frac{23.22}{41.8} \times \frac{1.34}{17.46} = 4.27 \times 10^{-3}$ g/h for
8.54×10^{-3} g/h 1.34.

2. Enrichment Factors $U^{234} : U^{235} = 27.8$
$U^{234} : U^{238} = 215$

3. Process Efficiency 6.0 %

CASE C. –

1.

Product - 100 g U-234

Deadline - six months

Assay - 50 %

Estimated cost -

Impact on other Isotope Programs

If above impossible -

Other products

Time

Approximate assay

Estimated cost

ABSTRACT

Magnetic separation of uranium to achieve high purity U 234 is described. Innovations included multiple stage operation, processing of small quantities of feed, and control of the inhalation hazard from alpha-active U 234. Starting with one per cent U 234 concentration, a two-stage separation program yielded 1.5 grams of uranium averaging 94 per cent U 234. This result was obtained through a U 234-U 235 enrichment factor of 36 at a U 234 process efficiency of 9 per cent.

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INTRODUCTION

As it occurs in nature, uranium exists as isotopes U 238, U 235, and U 234 with relative abundances, decay rates, and radiation energies tabulated below.*

Isotope	Abundance (%)	Half-life (yrs)	Radiation (Mev)
U 234 (UII)	0.0058	$2.35\text{-}2.67 \times 10^5$	α (4.76)
U 235 (AcU)	0.711	$7.07\text{-}8.8 \times 10^8$	α (4.56) α (4.40) γ (0.162)
U 238 (UI)	99.28	4.50×10^9	α (4.18)

For the purpose of achieving the physical isolation of U 235, the basic fuel element for atomic weapons, the mass-spectrographic method was developed for separating out the more abundant isotope, U 238, from the U 235. The third isotope, U 234, was neglected.

The magnetic separation to achieve high purity U 234 had a two-fold objective. First was the need to explore the potentialities of the mass-spectrographic method for processing limited quantities of rare isotopes in the heavy mass range. Associated with the U 234-U 235 separation was the determination of enhancement, production rate, and process efficiency factors which are helpful in predicting performance for other programs of a similar nature. Second, there was a specific requirement for U 234 in an enriched form for various research purposes. Up to this time, highly purified U 234 had not been produced by any process. Interest in enriched U 234 was partly due to the need for uranium isotopic standards for calibrating mass spectrometers. A request for top purity U 234 for cyclotron

* Nuclear Data, National Bureau of Standards Circular 499, p. 266, September 1, 1950

bombardment was outstanding. Also, availability of the pure isotope would simplify and refine measurements of U 234 half-life, alpha particle energy, spontaneous fission and other nuclear properties.*

Four different techniques applied to the problem of improving U 234 enrichment were tested using the mass spectrographs in the Beta Electromagnetic Plant. It was found that the intense uranium-ion beams which were produced by newly developed ion sources were not sufficiently resolved to achieve a high purity of U 234. Efforts toward improving the quality of resolution by limiting collection to well-focused ions were successful but necessarily reduced the utilized U^+ beam appreciably.

From preliminary studies with natural uranium run through the separators, it appeared that two separation stages connected in series would be more practical than attempting higher enhancements in a single stage at a severe cost to yield. It was anticipated that first stage processing would produce many grams of U 234 enriched to an intermediate level. Subsequently, second-stage separation of first stage produce was expected to isolate highly concentrated U 234 in gram quantities.

Very careful and precise adjustment of the mass-spectrographs was required to maximize U 234 enrichment relative to each of the other isotopes. This was possible through collector design, adjustment of resonance voltage controls, and operation techniques. The fourth method for improving the quality of separated U 234 was to recover selectively the material collected in the U 234 pocket. In this manner, it was possible to segregate material varying in U 234 concentration by several per cent.

* A number of shipments of enriched U 234 have been made, see Appendix A.

PRECEDING MAGNETIC SEPARATIONS OF URANIUM ISOTOPES

Uranium isotopic enrichment was first obtained by A. O. Nier in 1940.* The Nier spectrometer was a 7-inch radius, semi-circular instrument of the type used to measure relative abundance of isotopes. For 21 hours of operation with UBr_4 feed, metered U 238 current averaged 3×10^{-6} milliamperes. This corresponded to separate deposits of 1/2 microgram of enriched U 238 and 1/140 of this amount of enriched U 235. Based on resolution data, the natural uranium was enhanced 35 so that 20% U 235 purity was expected. U 234 could not be resolved from U 235 during this separation.

Previous U 234 Separation

Experimental production of U 234 in a modified Beta-size spectrograph was first tried by C. R. McKinney at Y-12 in 1946.** A total of 3.9 milligrams of the U 234 isotope was calculated to have been collected in 4 runs using 0.8% U 234 feed. The Beta machine had been revised to obtain the desired focus with an ion beam only 3° wide. The selected beam yielded U 238 currents of the order of one milliampere. Collector slots were one-half mass unit wide and the pockets were formed by stacking thin sheets of bent carbon. There was no determination of the isotopic abundance of the uranium extracted. The small amount recovered as product was insufficient for mass spectrometric measurement.

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* Nier, A. O., et al., Phys Rev 57, 546 (1940) "Nuclear Fission of Separated Uranium Isotopes."

** McKinney, C. R., The Production of U 234 by a Modified Calutron, D-4.250-30, May 1946.

Beta Mass Spectrograph, 1947

Mass spectrographs in the Beta Plant, used to separate the more abundant U 238 from the desired U 235, were of the 24-inch radius, semi-circular type, Figure 1. By 1947, ion currents of 110 ma of U^+ were established for each arc of the two-arc source unit. This corresponded to a deposit of one gram uranium per hour per arc, since practically all metered ions were retained. At prevailing feed vaporization rates of four grams uranium per hour per arc, process conversion efficiency of feed to separated isotopes was 25 per cent.

Resolution effected by the Beta magnetic isotope separator was such that two per cent of the U 238 ions present at the collector face crossed over to the U 235 product pocket, whereas one per cent of U 235 ions cross-contaminated the enriched U 238 material. This performance may be expressed in terms of three mass-unit enhancement as 100 for U 238:U 235 and 50 for U 235:U 238. Greater enrichment for U 238 concentrate than for U 235 product is attributed to several factors. The heavier U 238 ions travel a larger radial path and U^+ ions tend to lose kinetic energy by suffering collisions along their trajectory. Another advantage to U 238 enrichment accrues from monitoring both ion beams at their respective collector slots by maximizing U 238 ion current.

Difficulty in effecting complete isotopic separation arises from intense U^+ beams emitted by the ion source with an angular divergence of $\pm$ 12 degrees from the horizontal. Resolution between U 235 and U 238 isotopes for the 24-degree ion beam is aided considerably by use of magnetic shims along the ion path. However, the quality of resolution

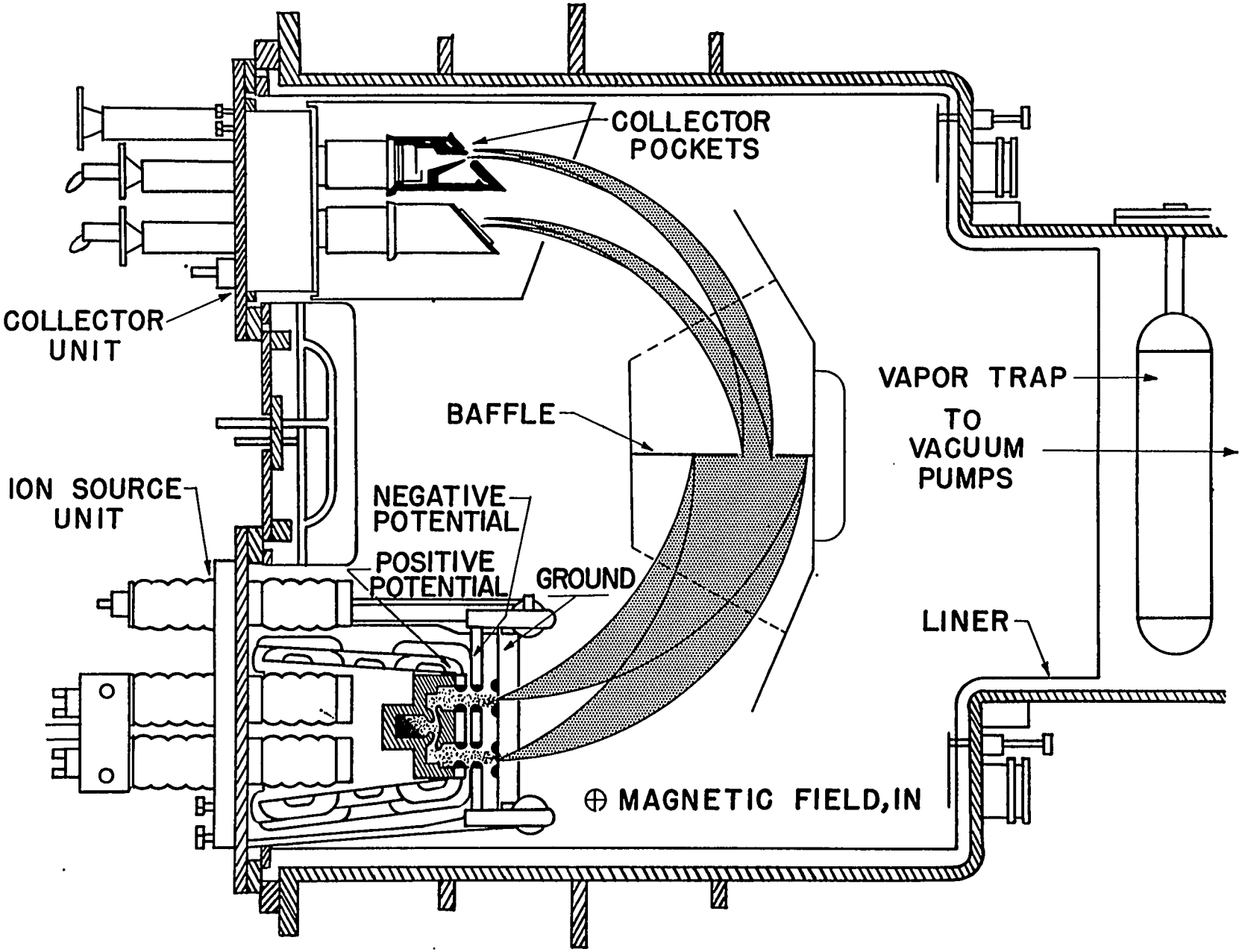


FIGURE I. BETA MASS SPECTROGRAPH

remains strongly dependent on the angular spread of divergent ion beams transmitted to the collector.

Collection of U 234 ions was not attempted on a production basis in the Beta Plant, since large gains could not be achieved with the small U 234 concentrations present in the feed. Also, about 70 per cent of separable U 234 ions were already trapped in U 235 product, due to the inappreciable one-unit-of-mass enrichment factor.

Summary

The mass spectrographs in the Beta Plant achieved U 234:U 235 enhancements of only 1.4, although the machine was potentially able to effect much better separation. The above performance was insufficient by a factor of 10 to 20 for obtaining high purity U 234. Experience with extremely narrow beams in Beta had not demonstrated adequate enrichment for cleanly separating U 234. Furthermore, by this method yields were curtailed to one milligram of uranium per run, whereas hundreds of milligrams were desired.

MASS SPECTROGRAPH FOR U 234 ENRICHMENT

The Beta mass spectrograph was adaptable to purification of uranium isotope U 234 through modification of geometry and use of refined operational methods.* Proposed changes in the separators, designed to increase enrichment, were initially tested on a small scale with natural uranium feed. Detailed adjustments of ion source geometry were required and performance parameters were measured to insure achievement of the desired result. Sufficient resolution of U 234 from U 235 required the stripping of about one-half of the U^+ beam produced by the U 235-U 238 separator. The indicated enhancement of U 234 with respect to U 235 was increased appreciably. Operational factors which tended to increase the width of focus, particularly on the low-energy side of the focal centerline, were reduced. An extremely stable and controllable arc was sought in order to avoid intermittent contamination of enriched U 234. Frequent observation of U^+ foci at the collector slots assisted in optimizing the focal width. The sensitivity of electrical controls was increased and the decline in U 234 purity due to high voltage sparking was minimized.

Ion Source

The ion source used in the U 234 program was similar in design and operation to the U 235-U 238 apparatus, Figure 2, except for details intended to emphasize enrichment. U^+ ions are formed in the source by sending 100 milliamperes of 150 volt electrons from a hot, 0.170" tantalum curved filament through a hollow graphite box containing UCl_4 vapor. Electrons emitted by the filament are collimated by the magnetic field and pass

* Hemphill, L. F. and Mitchel, G. W., (Eds.), The Electromagnetic Plant - 1949, Vol. II, "Established Operational Methods and Equipment," Y-AO-504.

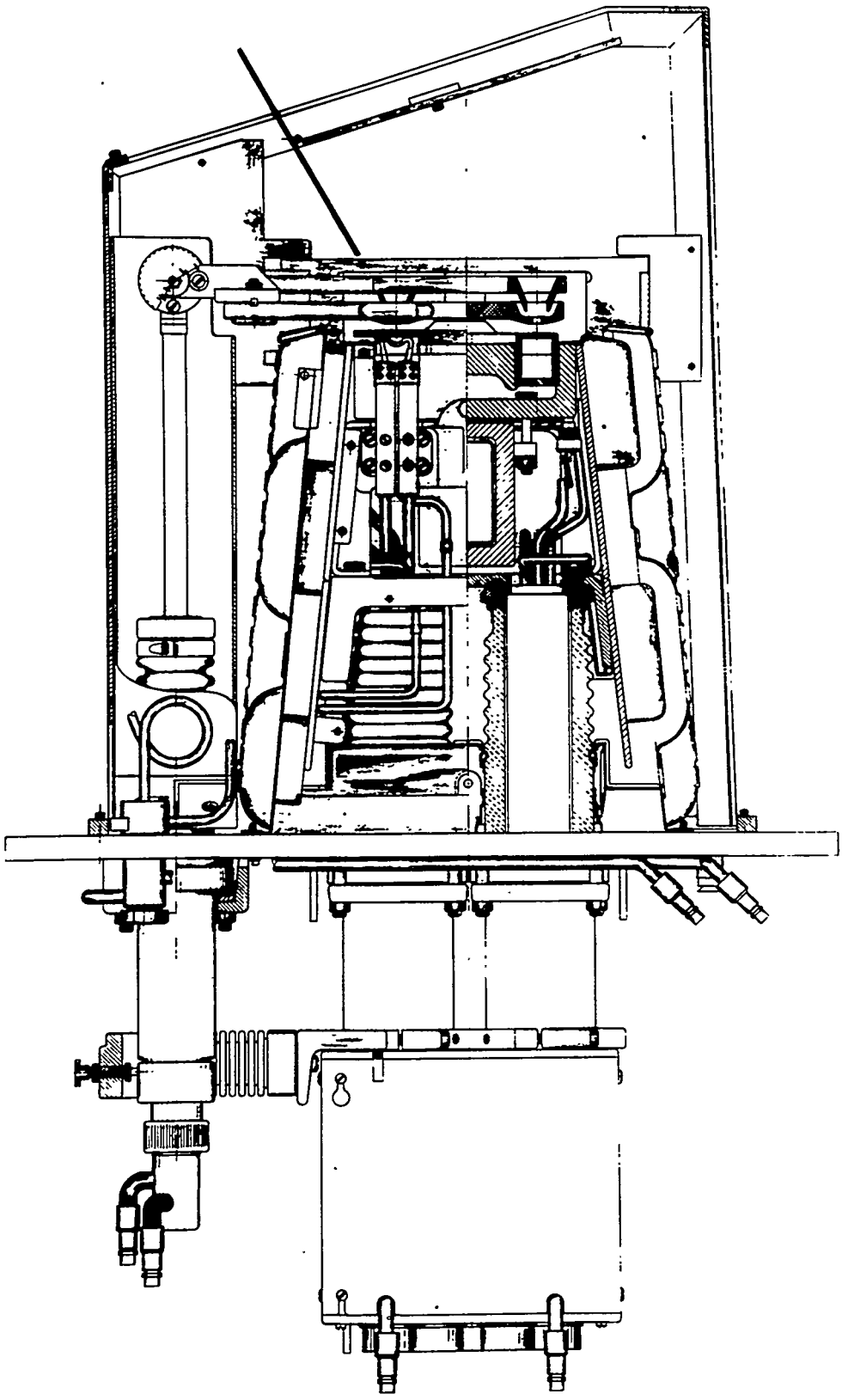


FIGURE 2. BETA ION SOURCE, MODEL M-12

through a defining slot curved to 3/8" radius, 0.078" wide and of 3/4" depth. At the end of the chamber opposite the filament, a similar defining slot is backed by an insulated graphite plate. This plate reflects electrons back into the ionizing region thereby improving efficiency.

Solid uranium compound is heated in the source unit to approximately 450° C in order to obtain suitable vapor pressures (~ 10 microns) to sustain an arc. The positive ions formed in the arc are drawn through a slit 7/16" x 6 13/16" and enter an accelerating region bounded by 37,000 volts positive at the ion exit and 38,000 volts negative at the accelerating electrode. The U^+ charges are therefore accelerated by a 75,000 volt potential in passing the 1/2" gap from the ionizing region to the accelerating slit of 5/8" aperture. Overall acceleration is, however, restricted to 37,000 volts by considerations which are determined by magnetic field and beam radius specifications of about 7000 oersteds and 24 inches respectively. Therefore, a decelerating carbon-slit electrode of 1 1/8" aperture at ground potential is positioned 1/2" away from the adjacent accelerating slit. Both the ion exit slit and the accelerating electrode designs have 15° bevels in the accelerating region.

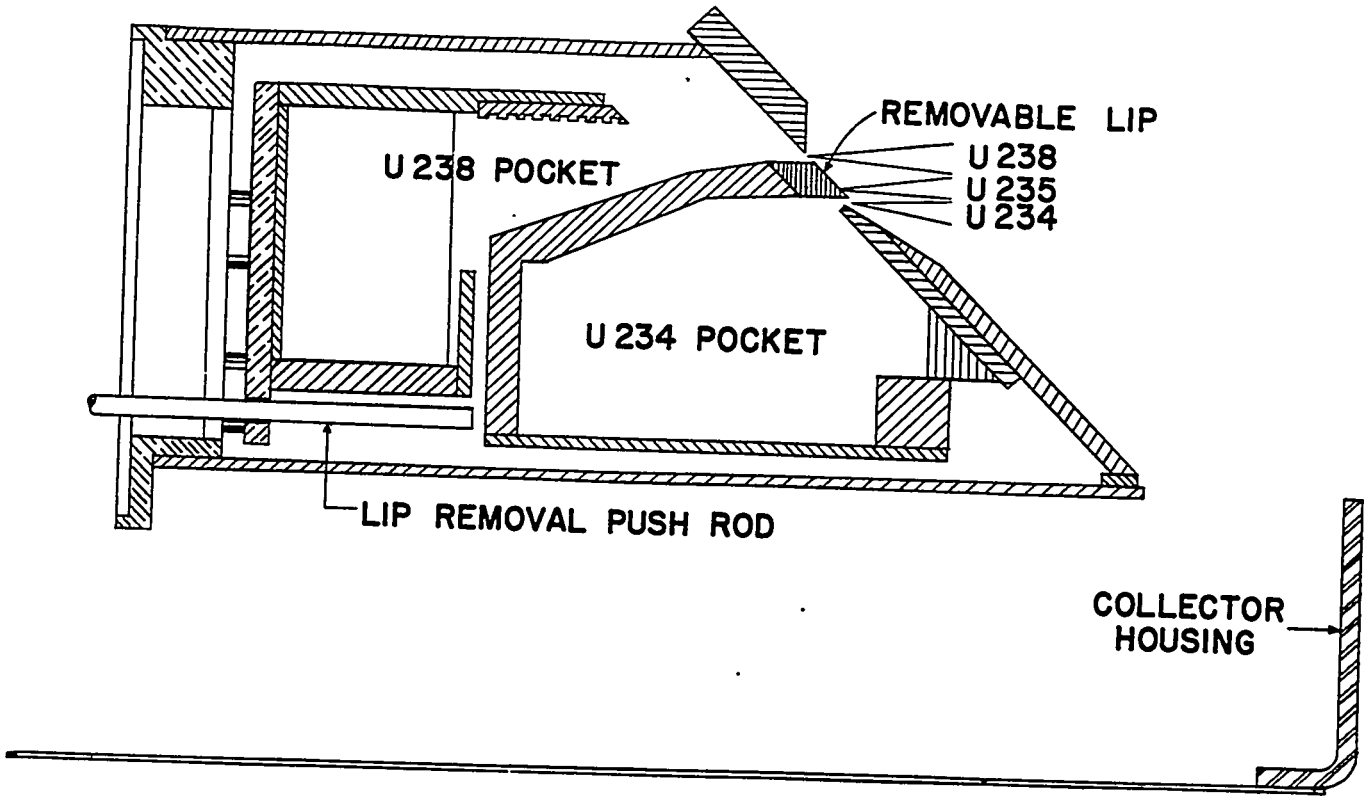
The source unit electrode geometry described above is identical for both stages of the U 234 program. However, the severe feed limitations of the second stage induced two modifications in source design. A single arc replaced the conventional twin arc of the first stage unit and a vapor valve mechanism was inserted as a fine control on UCl_4 vapor flow to the arc region.

Collector

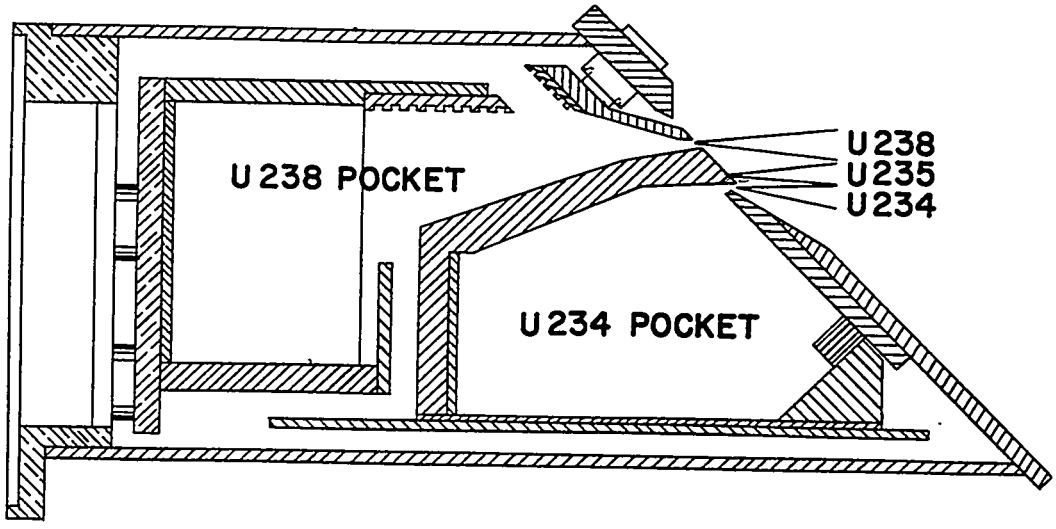
The useful features of Beta collectors for U 235-U 238 separation were extended to the production of high purity U 234. The all-carbon collector pockets, the monitoring devices and metal parts were incorporated in the U 234 program. However, U 234 collector design presented several new problems. First was the proper choice of ion entry slot width consistent with desired production rate and purity. Second was the design of durable slot boundaries, particularly between U 234 and U 235 ion foci.

The radius of curvature of the ion path fixes the distance between isotopic foci U 234 and U 235 at 0.1 inch, assuming infinitely thin focal widths. Due to space limitations, the decision was made to collect in both stages of the program only U 238 and U 234 ion beams. Since no U 235 slot was present in the collector, incident U 235 ions impinged on the carbon defining edge situated between the slots. The high kinetic energy of the U 235 ions caused appreciable wear, mainly 1/16" above the upper boundary of the U 234 slot. Continuing erosion of this member beyond 30 hours of separation destroys the barrier between the U 235 beam and the U 234 pocket. In the first stage collector, a removable carbon target was located at the U 235 beam focus to absorb U 235 bombardment during the initial half of the run, Figure 3. A mechanism was provided for moving the U 234 pocket assembly forward, thereby rejecting the removable section. In this manner the U 234 slot boundary was renewed. Due to the limited amount of feed available the second stage collector presented no durability problem.

The general problem of inter-relation of first and second stage U 234 enhancement relative to both U 235 and U 238 is considered in detail in Appendix B. If the accepted criterion is maximum U 234 purity from the



FIRST STAGE COLLECTOR



SECOND STAGE COLLECTOR

FIGURE 3. U 234 COLLECTORS

combined stages, then U 234:U 235 enhancement performance is the limiting factor. Both U 234 production and U 234 concentration of product enter into effective operation of the first stage whereas quality demands in the second stage are of paramount importance. It is possible through readily made collector slot changes to adjust output concentration inversely with volume.

U 234 Collector Geometry

	First Stage	Second Stage
U 238 Slot	0.100"	0.080"
Defining Edge	0.350"	0.363"
U 234 Slot	0.060"	0.050"

Angular Baffling

In the 180° Beta mass spectrograph, U^+ ions emerge from the source slit with an angular range of $\pm 15^\circ$ deviation about a plane normal to the slit. Ion orbits form an envelope of approximately uniform density about the zero degree ion path but tend to taper off beyond $\pm 10^\circ$. The widest section of the ion beam is at 90° where the width per degree is 7/16". The outermost ions of the envelope at this section issue from the source with large angular divergence. Despite magnetic shims, these particles broaden the image on the collector out of proportion to their respective intensity. To improve resolution, the ions of greater divergence were baffled out at the 90° point. A 12° angle beam (+7°, -5°) was selected as the most suitable geometry for the first stage separations. For the second stage of the program, the utilized ion beam was restricted to 8° (+5°, -3°). In addition, non-uniform baffle apertures were employed to screen out a segment of defocused ions in order to equalize focal widths along the 9" length of the collector slot.

U 234 PROGRAM - FIRST STAGE

This first stage was characterized by experimentation with U 234 collector slot width and angular baffle openings. Detailed changes in separator geometry were employed to determine various process performance combinations of quality and quantity . It was early discovered that although the relative feed concentration of U 238 to U 235 was 2.5 to 1, relative U 234 enhancements were of the order of 10 to 1 at a process efficiency of 10 per cent. The exclusion of low level increments from U 234 product assumes greater importance in this case.

Uranium Feed - First Stage

Beta recycle residue from the series operation of the Gaseous Diffusion Plant (K-25) and the Beta installation was used as the starting feed for the first stage of the U 234 program. Recycle uranium in Beta is the scattered, unresolved portion of vaporized feed which is recovered at the end of each run, and subsequently reconverted to UCl_4. Both K-25 and Y-12 had contributed toward U 234 enhancement in the Beta recycle. An adequate supply of accumulated Beta recycle was made available as feed for the U 234 program.

Enrichments resulting from both K-25 and Beta recycle operations in series are tabulated below. The U 235 component of K-25 product was enhanced 60 times while the associated U 234 component was enhanced 80-fold. The diffusion process does not attempt to strip the lighter constituent from enriched U 235. As a result, the level of U 234 was raised from 0.006% to 0.33% and, in addition, the ratio of 234:235 was increased one-third above the natural abundance ratio.

	Enhancement Factor for U 234			Concentration (%)		
Isotope	K-25	Beta	Total	Natural	K-25 Product	Beta Residue
U 238	80	4	320	99.28	69.7	70.4
U 235	1.33	4.3	6	0.711	30.0	28.3
U 234	----	---	---	0.0058	0.33	1.34

Beta separations of U 235 from U 238 did not attempt U 234 collection. The continued rejection of U 234 ions resulted in an increase in U 234 concentration in the Beta recycle material. This was particularly true during the 1946-47 period when K-25 production was no longer blended with the Beta feed bank.

The feed bank for the first stage U 234 program consisted of 4000 grams of uranium. Combined operations of K-25 and Beta recycling had raised the U 234 concentration to 1.34% and increased the ratio of 234:235 by a factor of six.

Uranium Throughput - First Stage

Uranium throughput for the first stage U 234 program amounted to 5600 grams of metal containing 75 grams of the desired isotope. The throughput was divided batch-wise into 14 parts. The rate of U 235 ion erosion markedly influenced the length of the individual run although substantially longer separations were obtained with the removable defining edge attached to the collector. Production time averaged 54 hours at vaporization rates of 3.7 grams uranium per hour per arc.

Run	Separation Period (hrs)	Uranium Vaporized (gms)	Uranium Vaporized (gms/hr/arc)	Throughput Mass Analysis U 234 (%)	U 234 (gms)	U 235 (%)	U 238 (%)
1	42	431	5.1	1.37	5.90	28.09	70.54
2	30	239	4.0	1.40	3.35	28.02	70.58
3	43	356	4.1	1.40	4.98	28.50	70.10
4	59	464	3.9	1.38	6.40	28.22	70.40
5	57	476	4.2	1.35	6.43	29.30	69.35
6	51	271	2.7	1.38	3.74	28.80	69.82
7	62	491	4.0	1.39	6.82	28.93	69.68
8	56	401	3.6	1.37	5.49	28.49	70.14
9	64	447	3.5	1.17	5.23	26.62	72.21
10	68	439	3.2	1.30	5.71	27.85	70.85
11	57	416	3.6	1.34	5.57	28.20	70.46
12	60	417	3.5	1.34	5.59	28.60	70.06
13	57	447	3.9	1.34	5.99	29.01	69.65
14	56	342	3.0	1.24	4.24	26.58	72.18
Total or Avg.	762	5637	3.7	1.34	75.44	28.25	70.41

Washing of unresolved uranium deposits from the surfaces of the mass spectrographs followed each separation run. The wash solution was then filtered, evaporated and adjusted for acidity prior to diethyl-ether extraction of the uranium. Conversion of the extracted material to the tetrachloride was made by a reaction at 120° for several hours with hexachloropropene as a chlorination reagent.

Uranium Production - First Stage

The 14 separation runs comprising the first stage U 234 program yielded a total of 18.5 grams of uranium assaying 40% U 234 and 48% U 235.

A survey of first stage production on a run basis reveals a strong inverse dependence between output and purity. Adjustments in separator geometry, such as U 234 collector slot width and angular baffle aperture, are mainly responsible for fluctuations in productivity and U 234 abundance.

Run	Product Uranium (grams)	Mass Analysis U 234 (%)	U 234 (grams)	U 235 (%)	U 238 (%)
1	0.799	41.68	0.333	46.18	12.14
2	0.717	43.81	0.314	39.08	17.11
3	1.319	39.20	0.517	45.56	15.24
4	1.270	42.35	0.538	42.83	14.82
5	1.450	42.77	0.620	45.62	11.61
6	0.466	46.57	0.217	40.99	12.45
7	1.570	37.54	0.589	50.35	12.11
8	2.140	34.71	0.743	53.06	12.23
9	1.270	46.04	0.585	40.11	13.85
10	1.580	40.56	0.641	45.07	14.37
11	1.760	32.82	0.578	54.90	12.28
12	2.080	35.71	0.743	53.33	10.96
13	0.971	44.24	0.430	44.59	11.17
14	1.110	42.79	0.475	44.44	12.77
Total, High Level	18.502	39.58	7.323	47.54	12.88
Total, Low Level	3.189	20.20	0.646	53.28	26.52

From past experience with collector processing, it was expected that uranium deposits on the carbon surfaces of the U 234 collector pocket would not be uniformly enriched. Surfaces adjacent to the slot opening are subject to greater cross-contamination. Also, enriched U 234 material which is not confined to the U 234 pocket during recovery operations tends to decrease in isotopic purity. In the process of extracting product level material, an attempt was made to segregate those segments of reduced enrichment. In this

manner, 3.2 grams of 20% U 234 material was obtained from the same 14 runs which produced 18.5 grams of 40% U 234, see Appendix C.

Practically all intermediate level U 234 concentrate was stockpiled in preparation for a second-stage separation. Small amounts of 52% and 46% U 234 material were withdrawn from the program to meet anticipated requirements of research laboratories. The 20% U 234 was not included in the second-stage feed.

Performance Standards - First Stage

On reporting performance of U 234 separator apparatus, consideration is given to both exclusion and inclusion of low level output. For the case where enrichment is paramount and it is desirable to exclude low level increments from U 234 product, U 234 process efficiency averaged 10% and U 234 enhancements relative to U 235 and U 238 are 18 and 162 respectively. If increase in U 234 production assumes greater importance, even at a small cost to purity, then first stage process efficiency is lifted to 11% associated with enrichments of 16 and 130.

The emphasis on enhancement reduced by fifty per cent the efficiency performance standards of which Beta equipment is capable, but multiplied the critical one-unit-of-mass separation factor 12-fold. The U^+ ion current averaged 70 milliamperes per arc for the two-arc ion source over a period of 760 innage hours. Concentrated U 234 production rates averaged 15 milligrams uranium per hour per arc.

Enhancement standards during the first stage U 234 separations were limited by production needs. Requirements of the consumer at this time indicated, however, the desirability of one attempt at maximum U 234 purity.

It was possible through use of a 0.040" U 234 slot width and a 6° angle beam to achieve further improvement in U 234 enrichment. This separation yielded 215 milligrams of 52% U 234 material from a single arc. The lesser quantity was due to a production rate of 5 mg U/hr/arc and the higher quality was determined by one and four mass unit enhancement factors of 30 and 207 respectively.

Run	U^+ Current (ma/arc)	U 234 Process Efficiency (%)	Enhancement	
			U 234:U 235	U 234:U 238
1	63	5.6	18.5	177
2	75	9.4	22.4	129
3	87	10.4	17.5	52
4	76	8.4	20.2	146
5	76	9.6	20.3	189
6	32	5.8	23.7	189
7	76	8.6	15.5	155
8	92	13.5	13.6	145
9	71	11.2	26.1	205
10	64	11.2	19.3	154
11	73	10.4	12.6	141
12	81	13.3	14.3	170
13	70	7.2	21.5	206
14	59	11.2	20.6	195
Average	70	9.7	17.6	162
Including Low Level Production		10.6	16.0	130

SECOND STAGE. - U 234 PROGRAM

The accepted criterion of second stage U 234 performance was maximum U 234 enhancement. Meanwhile, preliminary tests indicated uncertainties in separator behavior with charge weights limited to the 18 grams of uranium of intermediate enrichment. Consideration was given to use of purified U 238 as a "carrier" since 234:238 enhancement factors were sufficiently large. First stage enrichment performance standards were doubled in the second stage at a slight cost to yield and a peak purity of 96% U 234 was obtained.

Uranium Feed - Second Stage

Feed for the initial separation of the second stage was a composite of 18 grams of 39% U 234 and 18 grams of highly enriched U 238. There were two reasons for dilution of first stage U 234 product with 99.99% U 238 material. If U 238 feed concentration were sufficiently high for monitoring purposes, greater freedom in design of the U 234 collector pocket would result. The second, and more important, consideration was maximizing U 234 product purity. The feed limited situation motivated a preliminary study of enhancement versus feed weight. It was possible to evaluate potential gains in enhancement performance for various U 238 additives to first stage output. These gains are achieved through a reduction in U 234 feed concentration. Of the several variations in feed size tested, the objective of maximum U 234 purity was achieved with a 36 gram composite.

	Medium Concentrated U (grams)	Mass Analysis					
		U 234		U 235		U 238	
		(%)	(gms)	(%)	(gms)	(%)	(gms)
	0.004	51.7	0.002	35.7	0.001	12.6	0.001
	0.600	46.0	0.276	40.1	0.241	13.9	0.083
	0.794	44.2	0.351	44.6	0.354	11.2	0.089
	1.025	42.8	0.439	44.4	0.455	12.8	0.131
	6.224	41.4	2.577	44.3	2.757	14.3	0.890
	1.503	40.6	0.610	45.1	0.678	14.3	0.215
	4.135	35.8	1.480	51.5	2.130	12.7	0.525
	1.976	35.7	0.706	53.3	1.053	11.0	0.217
	1.675	33.8	0.566	54.9	0.920	11.3	0.189
Total or Avg.	17.936	39.1	7.007	47.9	8.589	13.0	2.340
Carrier	18.000	0.0	0.000	0.01	0.002	99.99	17.998
Composite	35.936	19.50	7.007	23.91	8.591	56.59	20.338

Blending of U 238 concentrate with partially enriched U 234 yielded a starting material assaying 20% U 234 and 24% U 235, of which the U 234 constituent amounted to 7 grams.

Uranium Throughput - Second Stage

Separation of gram quantities of highly concentrated U 234 isotope involved three recycles of the original 36 gram feed bank, and a fourth recycle purely for recovery purposes at termination of the program. Integrated uranium throughput for the four second-stage separations amounted to 133 grams of metal containing 23 grams of U 234. The quantity of feed available for electromagnetic reprocessing depended mainly on recovery efficiency and to a smaller extent on enriched U 234 yields. Following the first pass, an average of one-fifth of the U 234 feed was diverted to holdup on equipment. U 234 concentration of the feed decreased after the initial separation as a result of U 234 product extraction and other uranium pickup from process equipment. Although the beginning U 234 level was 19.5%, total throughput averaged only 17.5% U 234.

Run	Separation Period (hrs)	Uranium Vaporized (gms)	(gms/hr/arc)	Throughput Mass Analysis U 234 (%)	U 234 (gms)	U 235 (%)	U 238 (%)
1	9.0	36.0	4.0	19.50	7.007	23.91	56.59
2	9.0	31.6	3.5	18.27	5.777	23.25	58.48
3	13.5	33.5	2.5	16.83	5.635	22.72	60.45
4	10.3	31.9	3.1	15.04	4.802	22.34	62.62
Total or Avg.	41.8	133.0	3.2	17.46	23.221	23.08	59.46

Production time averaged 10 hours per run at vaporization rates of 3.2 gm U/hr for the single arc ion source. Scattered U^+ ions and non-ionized uranium deposits were partially recovered by acid spraying the machines in an enclosed stainless steel bin prior to disassembly and by scrubbing of unit parts. The spray washing technique also reduced the considerable inhalation hazard in handling alpha-active U 234 in dry form.

Uranium Production - Second Stage

The second stage U 234 program separated 1.5 grams of highly enriched U 234 in 42 hours of mass spectrograph operation. The isotopic mixture averaged 94% U 234 and 4.5% U 235. U 238 contamination in product was one-third the U 235 level. The third run was not only the most productive but also yielded the highest purity, 96% U 234.

A comparison of U 234 output per run must consider such factors as variable mass spectrograph performance and changes in U 234 feed weight. The breakdown of ion source components during the initial two separations reduced productivity as well as purity. The final two runs produced 1 gram of 95% U 234 under more normal conditions. Productivity was expected to decline with the number of elapsed runs due to partial recovery of unresolved uranium and to enriched U 234 leaving the process. Because of the absence of U 234 make-up, the feed bank is depleted thereby reducing potential output.

Run	Product Uranium (grams)	Isotopic Analysis U 234 (%)	U 234 (gms)	U 235 (%)	U 238 (%)
1	0.099	89.34	0.088	7.72	2.89
	0.078	89.35	0.069	7.74	2.91
2	0.284	94.00	0.267	4.45	1.53
	0.040	89.35	0.036	7.74	2.91
3	0.378	95.99	0.363	3.02	0.98
	0.151	94.83	0.144	3.99	1.19
4	0.345	94.70	0.327	4.02	1.28
	0.102	92.73	0.095	5.87	1.39
Total	1.477	94.04	1.389	4.47	1.49

Selective recovery techniques used in processing the U 234 collector pocket made it possible to segregate product batches varying in U 234 concentration. From each separation a higher purity batch was extracted in addition to a somewhat lower level segment of smaller size. The precision of the Y-12 Mass Spectrometer Laboratory is such that a 0.1% limit of error is assigned to each of the isotopic measurements in the product.

Performance Standards - Second Stage

Evaluation of mass spectrograph performance is based upon process efficiency and enhancement parameters obtained during the latter part of the second stage U 234 program. Approximately 52 grams of UCl_4 were vaporized in each separation run. Metered U^+ ion current averaged 36 milliamperes which is well below the 70 milliamperes established in the first stage. Apparently there is a tendency towards lower production rate as well as reduced vaporization rate with gram quantities of feed. This effect is indicated by U 234 process efficiencies of 10.6% and 8.9% for the first and second stages.

Second stage enrichment requirements entered into narrowing the U 234 collector slot width to 0.050" and baffling the U^+ ion beam to 8^o. Through use of higher resolution geometry and refinement of operational methods, the first stage 234:235 enhancement standard of 18 was increased to 36. Likewise, the 234:238 enhancement factor was lifted from 160 to 300. The batch of maximum enrichment contained 380 milligrams of uranium concentrated to 96% U 234. Enhancement parameters associated with the top purity material are 43 for 234:235 and 350 for 234:238.

Since the U 234 program required no major changes in Beta mass spectrographs or operational techniques, one can now make estimates for all uranium isotopic separations based on U 234 performance.

Run	U^+ Current (ma/arc)	U 234 Process Efficiency (%)	Enhancement 234:235	Enhancement 234:238
1	27	2.3	13.8	92
2	25	5.2	24.1	194
3	37	9.0	40.2	301
4	35	8.8	31.2	293
Average	32	6.0	27.8	215

Uranium Recovery - Second Stage*

Chemical processing in the second stage was governed largely by the limited supply of intermediate level U 234. Following each run the unresolved portion of vaporized uranium was recovered as completely as possible and reconverted to new feed. In assessing U 234 recovery efficiency, attention is paid to recovery between runs as well as to the terminal recovery.

Between separation runs, about four-fifths of unresolved uranium was recovered and converted to UCl_4. The diversion of 20% of unseparated U 234 to holdup on equipment reduced production by a proportionate amount. On completion of the isotope separation phase of the program, the recovered residue of unresolved uranium totaled 41 grams of metal assaying 13.5% U 234 and 22% U 235. This material is available for replenishing the supply of highly enriched U 234.

A large fraction of the original 7 grams of U 234 in the feed was processed four times in the mass spectrographs. Of the 23 grams of U 234 throughput, only 0.1 gram was unrecovered. Recovery efficiency evaluated

*Case, F. N., Facilities for Processing Alpha-Active Isotopes, Y-659. Sept. 1950.

in terms of initial feed and throughput are 98.7% and 99.6% respectively.

Recycle recovery performance is based on accounting for U 234 isotope. The advantage to this method lies in the high degree of U 234 impoverishment in pickup uranium. Pickup from previous use of the equipment amounted to 7 grams of 92% U 238 with 8% U 235.

HEALTH HAZARDS - SECOND STAGE

The four passes through the separation process represented a recycle throughput of 133 millicuries and a product output of 8 millicuries of 4.8 Mev alpha particles from the U 234. Non-selective recovery from the ion source and liner jacket and selective recovery from the collector followed each separation. Due to the alpha-inhalation hazard associated with processing U 234 in dry form, the material was handled in such a manner as to reduce airborne dispersion.

Uranium chlorination was followed by washing the UCl_4 into a stainless steel bottle with carbon tetrachloride and outgassing the material in a vacuum furnace. Towards the latter part of feed preparation, there is a transition from wet chemistry to the dry state. Sealing the feed bottle and charging the source unit are dry operations.

On completion of a run, the mass spectrograph was removed from the magnet and openings in the liner promptly sealed. The collector unit was then transferred from the liner jacket to a plastic glove box for disassembly and removal of the U 234 product pocket, Figure 4. These exposures were of short duration so that personnel protection was confined to use of respirators.

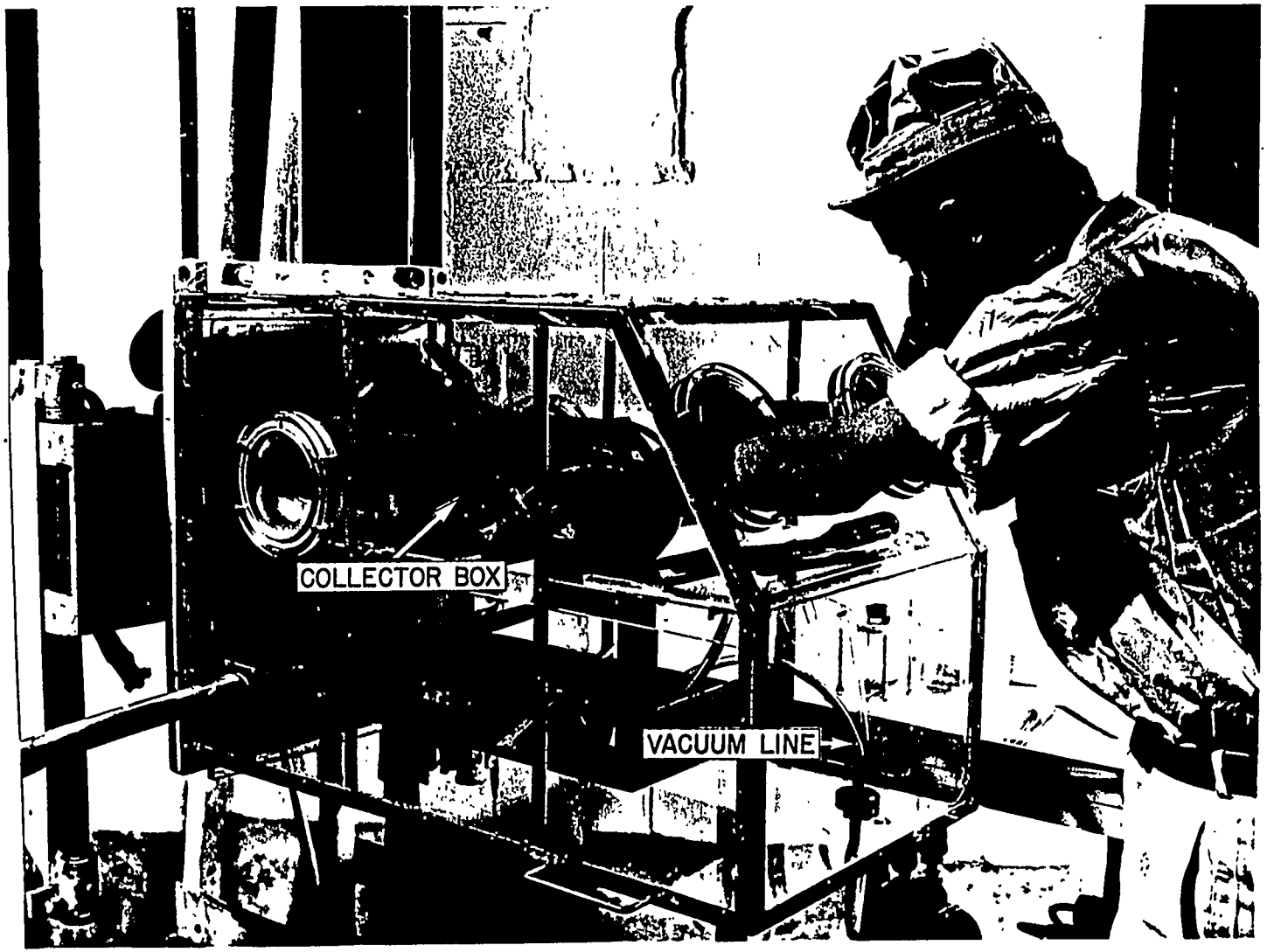


FIGURE 4. PORTABLE GLOVE BOX

The carbon U 234 pocket was transferred to a ventilated chemical hood provided with glove ports where vacuum-scraping techniques were used for extracting enriched U 234, Figure 5. Leaching, muffling and purification of the extracted uranium were performed in the same location. Respiratory protective devices were used as a secondary safeguard during product processing.

Recovery of adherent material from the surfaces of the ion source and liner involved considerable agitation for an extended period. Consequently, this operation was totally confined in a spray washer until the dry oxides of uranium were either dissolved in heated nitric acid solution or at least thoroughly wet down, Figure 6. Subsequent disassembly and intensive cleaning of unit parts prior to reconstructing the machine were performed with respirators as the primary safeguard.

Recycle processes, consisting of filtration and evaporation of approximately 50 gallons of wash solution, were performed in closed systems. Extraction and precipitation of uranium from concentrated wash and muffling of contaminated graphite were carried on in high-air-velocity hoods.

Health physics monitoring techniques included: air sampling in work areas, respirator filter analyses, and twenty-four-hour urine specimens. Data on personnel engaged in potentially hazardous activities were correlated with monitoring measurements to determine exposures corresponding to individual operations. To date, excretion samples from only the first run have been analyzed, Appendix D. Results indicate that by enclosure of contaminated units in acid spray tanks, glove boxes and glove hoods, the bulk of dry U 234 was contained. However, exposures were above the permissible level for continuous plant operation.

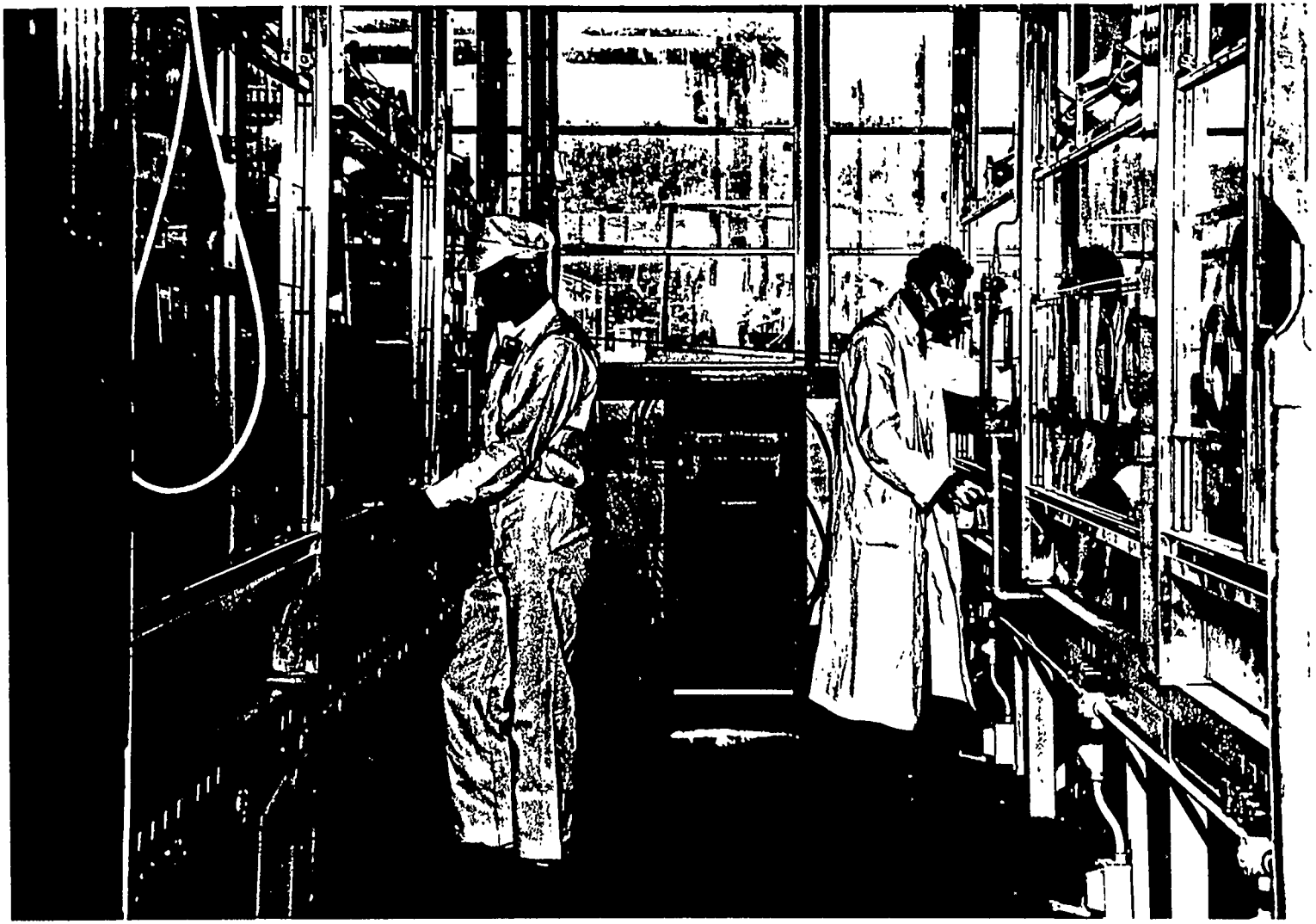


FIGURE 5. HOODS FOR HANDLING ALPHA ACTIVE MATERIALS

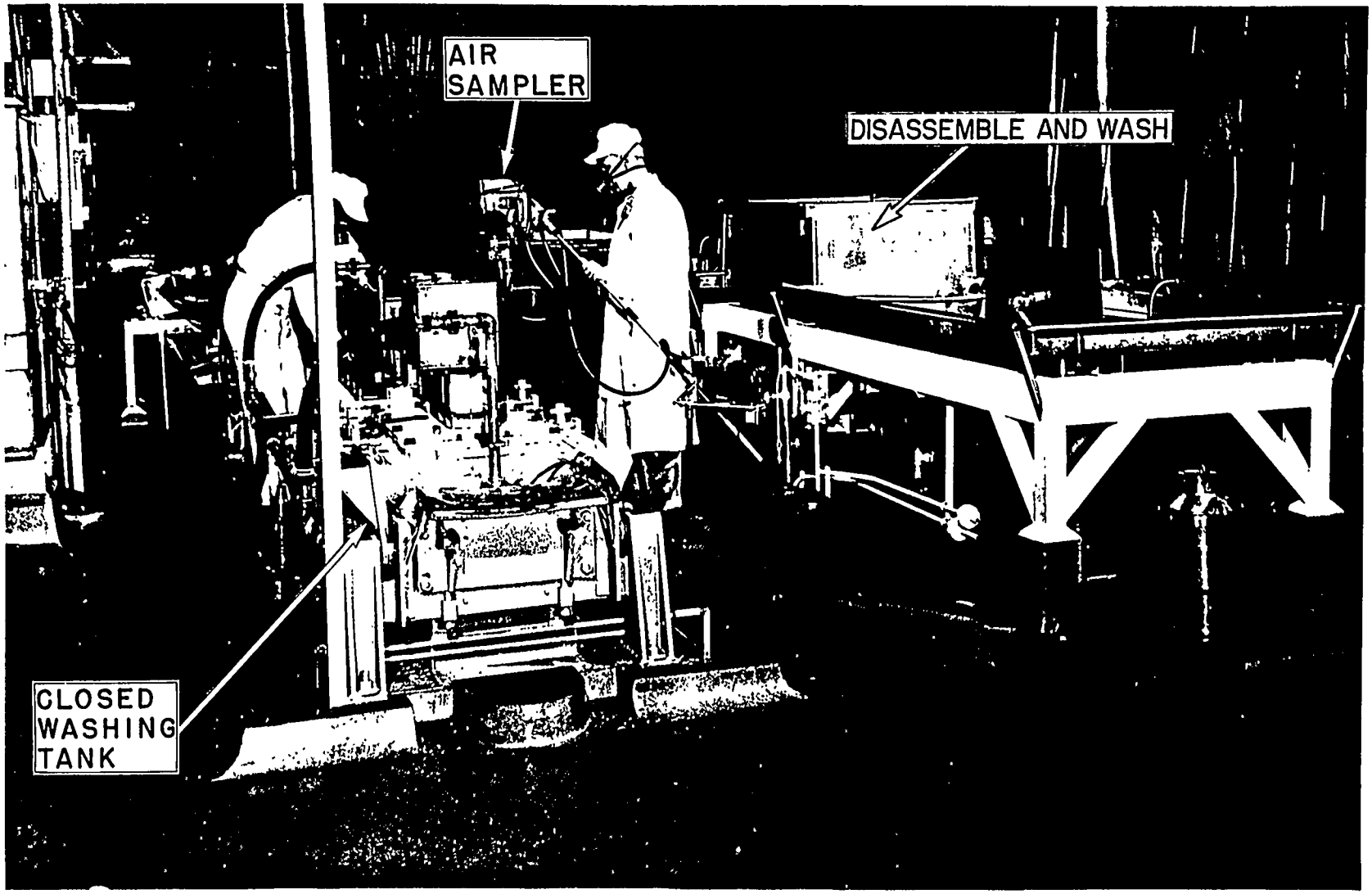


FIGURE 6. ISOLATED RECOVERY AREA

CONCLUSIONS

The potentialities of the electromagnetic process for the separation of heavy isotopes differing by one unit of mass have been demonstrated in the concentration of U 234 to better than 94% purity.* The four methods used for achieving highly concentrated U 234 included high resolution separator geometry, multiple stage processing, selective extraction of product, and operational techniques which favored enrichment of U 234 with respect to the heavier isotopes. The overall enhancement of the 96% U 234 sample, the product of the several single stage enhancement factors, is calculated to be 760 and 57,000 for 234:235 and 234:238 respectively. If one includes reduction in quality of second stage feed due to dilution with U 238 and depletion of U 234, one and four mass unit enhancements of initial feed are 670 and 5150. Finally, the purest U 234 was enhanced above natural abundance by factors of 4000 and 1,640,000 with respect to the U 235 and U 238 constituents.

* Livingston, R. S., and Shipley, E. D., Catalog of Highly Purified Uranium Isotopes, Y-614, June 1950.

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Personnel assisting in establishing the feasibility of separating U 234 included J. M. Googin, T. P. Sprague, and C. C. Sharp, all of whom performed collector disassembly, enriched U 234 extraction, and subsequent product purification. Scattered unresolved uranium was recovered from unit surfaces and converted to feed material under the supervision of J. S. Reece and F. N. Case. H. C. McCurdy did the necessary engineering and procurement for the program and aided in preliminary testing of the electromagnetic equipment. Analytical and mass analysis laboratory services were supplied by L. A. Stephens and R. F. Hibbs, respectively. Measurements of alpha activity in the air and in excretions were performed by the health physics section under E. G. Struxness. Mrs. B. Buckminister performed the statistical computations incident to the program.

APPENDIX A: SHIPMENTS OF ENRICHED U 234

	Uranium (mgs)	Isotopic Composition U 234 (%)	U 235 (%)	Proposed Use
University of California Radiation Laboratory	10	95.99	3.02	Cyclotron bombardment
Los Alamos Scientific Laboratory	10	94.00	4.45	Counting standards
K-25 Laboratory	30	94.00	4.45	Uranium assay
Argonne National Laboratory	1.5	95.99	3.02	Spontaneous fission
Argonne National Laboratory	5	95.99	3.02	Half-life
Argonne National Laboratory	10	89.35	7.73	Half-life
Argonne National Laboratory	10	51.7	35.6	Half-life
Argonne National Laboratory	10	46.0	40.1	Half-life
University of California Radiation Laboratory	2.6	46.0	40.1	---
Y-12 Physical Analyses Laboratory	100	44.2	44.6	Spectroscopic isotope shift
Brookhaven National Laboratory	2	51.7	35.6	---
Oak Ridge National Laboratory	0.005	51.7	35.6	Alpha range standards
Hanford Operations	10	95.99	3.02	---
Fissionable Standard Samples Committee	844	93.63	4.83	Mass spectrometer standards

APPENDIX B: THEORY AND COMPUTATION

U 234 isotopic enrichment is calculated for the case of the two-stage magnetic separator where initial stage product provides second stage feed. There are two uranium contaminants to be minimized - isotopes 235 and 238 which are separated one and four mass units, respectively, from U 234.

Given:

$w_o + x_o + y_o$	as	U 234,5,8 grams of feed respectively
$w_p + x_p + y_p$	as	U 234,5,8 grams of product, first stage
$w'_p + x'_p + y'_p$	as	U 234,5,8 grams of product, second stage

By definition:

	Enhancement	
	one mass unit	four mass units
First Stage	$N_{wx} = \frac{w_p/x_p}{w_o/x_o}$	$N_{wy} = \frac{w_p/y_p}{w_o/y_o}$
Second Stage	$N'_{wx} = \frac{w'_p/x'_p}{w_p/x_p}$	$N'_{wy} = \frac{w'_p/y'_p}{w_p/y_p}$

Then weight per cent U 234, first stage product, is derived in terms of initial feed and enhancement factors as:

$$\frac{w_p}{w_p + x_p + y_p} = \frac{w_p}{w_p + \frac{w_p}{N_{wx}(w_o/x_o)} + \frac{w_p}{N_{wy}(w_o/y_o)}} = \frac{w_o}{w_o + \frac{x_o}{N_{wx}} + \frac{y_o}{N_{wy}}}$$

Likewise weight per cent U 234, second stage product, is calculated to be:

$$\frac{w'_p}{w'_p + x'_p + y'_p} = \frac{w'_p}{w'_p + \frac{w'_p}{N'_{wx}(w_p/x_p)} + \frac{w'_p}{N'_{wy}(w_p/y_p)}} = \frac{w_o}{w_o + \frac{x_o}{N_{wx}N'_{wx}} + \frac{y_o}{N_{wy}N'_{wy}}}$$

APPENDIX C: U 234 LOW LEVEL PRODUCTION - FIRST STAGE

Uranium (grams)	U 234 (%)	U 234 (grams)	U 235 (%)	U 235 (grams)	U 238 (%)	U 238 (grams)
0.213	32.49	0.069	42.97	0.092	24.54	0.052
0.038	32.05	0.012	46.80	0.018	21.15	0.008
0.052	29.99	0.016	41.28	0.021	28.73	0.015
0.085	27.30	0.023	48.39	0.041	24.31	0.021
0.208	26.47	0.055	55.62	0.116	17.91	0.037
0.482	21.64	0.104	39.32	0.190	39.04	0.188
0.765	20.44	0.156	49.74	0.381	29.82	0.228
0.143	19.27	0.027	57.86	0.083	22.87	0.033
0.630	18.80	0.118	54.71	0.345	26.49	0.167
0.497	12.20	0.061	72.79	0.362	15.01	0.074
0.085	5.94	0.005	65.27	0.055	28.79	0.025
3.198	20.20	0.646	53.28	1.704	26.52	0.848

APPENDIX D: HEALTH MONITORING DATA - RUN #1

Operation	Operation Interval (min)	Airborne Concentration** Gen'l Room (α/min/M^3)	Airborne Concentration** Work Level (α/min/M^3)	Personnel Respirator Fore Filter (α/min)	Personnel Respirator Aft Filter (α/min)	U in Urine*** (α/min/24 hr)
Feed Preparation						
1. Wet Chemistry	25	69	176	198,000	0	662
2. Dry Chemistry	16	129	15,082*	565	0	307
Unit Removal						
1. From Magnet	5	324	---	6	0	29
2. Collector Removal	2	4	1011	15	0	5.6
Collector Processing						
1. Pocket Removal (Gloved Box)	21	223	356	33	20	8
2. Pocket Scraping (Hood)	127	85	0	22	0	116
3. Product Chemistry	395	17	132	107	33	16
Source and Liner Processing						
1. Source and Liner Spray	204	102	---	377	48	77
2. Source Disassembly and Wash	94	19,400	163	111	9	59
Recycle Processing						
1. Carbon Muffling	39	0	0	42	11	47
2. Machine Wash Extraction	--	--	--	475	55	216

* UCl_4 spill on sealing the feed bottle

** Maximum permissible amount of uranium in air for continuous plant operation is limited to 70 α's/min/M^3.

*** Maximum permissible amount of insoluble uranium present in the body is indicated by 70 α's/min per 24 hour urine specimen.